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FABRICATION OF HIGHLY DENSE Si_3N_4 CERAMICS WITHOUT ADDITIVES BY HIGH PRESSURE SINTERING

K. Takatori, M. Shimada, and M. Koizumi

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16. Abstract

Silicon nitride (Si_3N_4) is one of candidate materials for the engineering ceramics which can be used at high temperatures. The mechanical strengths of hot-pressed or sintered Si_3N_4 ceramics containing some amount of additives, however, are deteriorated at elevated temperatures. To improve the high temperature strength of Si_3N_4 ceramics, an attempt to consolidate Si_3N_4 without additives was made by high pressure sintering technique. Scanning electron micrographs of fracture surfaces of the sintered bodies showed the bodies had finely grained and fully self-bonded sintered bodies were 310N/m^2 at room temperature and 174N/m^2 at 1200"C.

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FABRICATION OF HIGHLY DENSE Si_3N_4 CERAMICS
WITHOUT ADDITIVES BY HIGH PRESSURE SINTERING

Kazumasa Takatori, Masahiko Shimada[1] and Mitsue Koizumi

The Institute of Scientific and Industrial Research, Osaka University: Yamadaoka, Suita-shi 565 Japan

1. Forward /*1506

Great attention has been paid to silicon nitrade in the field of natural resources and energy, as one of many candidate materials for ceramics engineering which can be used at high temperatures.[-2] Silicon nitrade is expected to be one of candidate materials because of the following strong points: 1) both silicon and nitrogen exist as abundant earth resources; 2) in the domain of very high temperature which exceeds the existing maximum usage temperature for the metal materials (i.e., 1100°C) silicon nitrade maintains its strength; 3) its specific gravity is about 3.2 which is about 1/3 of super alloy; 4) it has superior corrosion resistance; 5) since its coefficient of heat expansion is very small and thus, is hardly subjected to plastic deformation, it is suitable for precision parts; 6) it has superior heat shock resistance. Taking advantage of these strong points, the use of silicon nitrade as the parts for high temperature operating machines, furnace material, precision parts or bearings has been under consideration.[3]

Ceramics is generally produced by the sintering method, but it is well known that silicon nitrade is hard to be sintered. Precise sintered body cannot be obtained by an ordinary method. As a result, at present, to produce the precise sintered body of silicon nitrade several percentages of oxide (Y_2O_3, Al_2O_3, MgO) are added andthe hot-press or sintering under normal pressure methods are used.[4-6] The silicon nitrade sintered body obtained by the above methods is a very strong material with more than $10N/m^2$

*Numbers in the margin indicate pagination in the foreign text.

flexural strength in the temperature between room temperature and' less than about 1000°C. However, added oxide separates out and exists as glass or the second phase of the crystal. As a result, it deteriorates the expected high temperature strength of silicon nitrade at over 1000°C

As the silicon nitrade sintered body which has less mechanical strength deterioration, one without any additives is prefered, but there has been very little research dealing with the production of precise sintered body which is usually described as the material with sintering difficulty by applying high pressue when sintered, and have been studying the sintering organization and the heat-mechanical characteristics of the sintered body.[8] In the research, we have obtained highly dense silicon nitrade ceramics without additive by high pressure sintering which indicates very low deterioration of the strength under high temperature. Here is the report on this silicon nitrade sintered body.

2. Method of the Experiment

As the high pressuring sintering, a cubic model high pressure generating device with its tip of 15mm was used. The conditions were 5.0 GPa, 1800'C, less than 120 minutes pressure, temperature and time. As the starting material high density α-powdered Si_3N_4 was used.[9] It was then cold formed at 100MPa through the gold mold, and after that a 5ϕ x 2.5t mm^3 pellet was obtained. The density of the formated body was about 40% of the theoretical Si_3N_4 density. The Archimedes method was used to measure the volume density of the Si_3N_4 sintered body obtained by high pressure sintering.

In Si_3N_4 there exists two crystal phases: α-phase and β-phase.[10] The α-phase changes to the β-phase in the temperature domain of more than 1500°C when additives are added and hot-pressured.[11] In this study, in order to research its behavior when phase is

changed at the time of sintering without any additives and also the effect of the phase change to its precision, a quantitative analysis of the crystal phase was exercised using the powdered X-ray diffraction method on the starting material and the sintered body.[12)]

The mechanical characteristics of the sintered body was evaluated by examining the temperature dependency of microhardness. The high temperature Vickers microhardness measuring device was used to measure the microhardness in the temperature domain between room temperature and 1200°C with 200g load. The microstructure of the inside of the sintered body was observed using SEM.

3. Experiment Result and Observation

Under several conditions, the silicon nitrade high pressure sintering experiment was conducted. As the result of measuring the precision and existing ratio of the crystal phase of the obtained sintered body, and the observation of the microstructure, the following results were obtained.[13)]

1) β-phase is very stable under high temperature and high pressure. As the operation pressure increased, the changing temperature from the α-phase decreased.

2) Si_3N_4 sintered body which has almost theoretical density was obtained by 1.0GPa, 1600°C, 200 minutes high pressure sintering.

3) When α- and β-Si_3N_4 were used as the starting materials, /1507 there was no difference in its minute behavior observed.

4) The fractured surface of the sintered body indicates the inner particle destruction, and therefore, it is assumed that active intraparticle self-bonding is done.

5) The Vickers microhardness of self-bonded sintered bodies were $310N/m^2$ (200g load) at room temperature in the vacuum condition.

Figure 1 shows the SEM of the fracture surface of the sintered

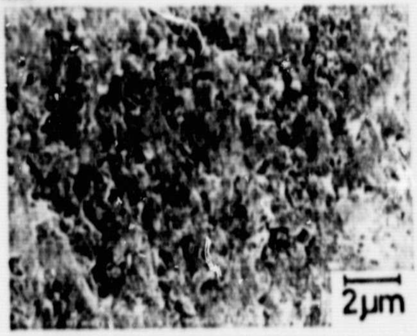

Fig. 1 Microphotographs of the fracture surface of the highly dense Si_3N_4 sintered body

body obtained by 3.0 GPa, 1800°C and 5 minutes treatment. The volume density of this sintered body was 99.8% of the theoretical density. β-phase content rate was 4wt% with the starting material, while it was 88wt% with the sintered body. The microstructure of the inside of the sintered body, as obvious from Figure 1, shows the formation of the polyhedron of the microparticles which are less than 1μm. Since the microparticles are closely self-bonding among particles, it is assumed that the great deterioration of the Si_3N_4 sintered body without additives at high temperature is very small.

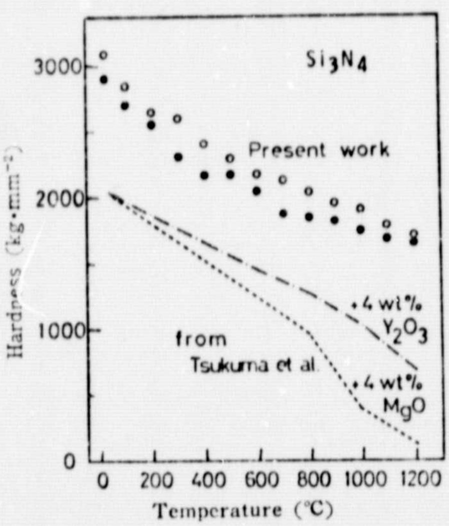

Fig. 2 Temperature dependence of microhardness of the highly dense Si_3N_4 sintered bodies

○ : 3.0 GPa, 1600°C, 30 min
● : 3.0 GPa, 1800°C, 30 min

Figure 2 shows the temperature dependence of the Vickers microhardness of the non-additive high density Si_3N_4 sintered body obtained by this research. In this figure the recorded ratios [14] of the microhardness of the Si_3N_4 sintered body which contains Y_2O_3 or MgO, used as the sintering aiding agents in the hot-press process, are also shown. Compared with the sintered body with additives, the Si_3N_4 sintered body without additives indicates greater microhardness in the whole

temperature domain measured, especially, there is no salient deterioration of microhardness at more than 800 - 1000°C.

4. Conclusion

As to Si_3N_4 which is expected to perform well as the high temperature high strength material, the existing hot-press sintering body using sintering aiding agents has a problem of deteriorating its mechanical performance at high temperature. In order to improve the above problem, Si_3N_4 sintering without additives was successfully made using the high pressure sintering technique. The non- additive high density sintering body has relatively similar size particles, and microhardness did not deteriorate much at the temperature of more than 1000°C, thus, it is assumed to be the superior material which can maintain its strength even in high temperature.

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